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ORIGINAL CONTRIBUTION

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Studies on the Mayak nuclear workers: dosimetry

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Background and aim of the study

From the onset of the Mayak Production Association (Mayak PA), the key task of specialists at the Radiation Safety Department of the enterprise and scientists of Branch No.1 of the Biophysics Institute (FIB-1)[1] has always been to arrange individual dosimetry monitoring of the personnel. In the 1940s and 1950s it was only possible to measure external exposures. A corresponding program was introduced in June, 1948 and since then all workers have been under individual external exposure monitoring.

The situation of monitoring internal exposures was more intricate. During the early years of the enterprise operation, primary attention was focused on the exposure to ^{90}Sr and ^{137}Cs. Assessments of body burdens of beta activity due to uranium fission, based on ^{137}Cs measurements in the period 1977–1995 showed, however that these activities were well below permissible levels (Fig. 1). It took several years to realise that the major risk for personnel health originates from internally deposited plutonium. Although first attempts to assess plutonium body burdens were made in 1953, it was not until 1970 that it became possible to implement a set of techniques for measuring plutonium in biological material and to develop a biokinetic model of plutonium transport and excretion in the human body [1]. Assessments based on these methods revealed an excess of permissible levels in many cases (Fig. 2).

In this paper, the term OLD_DOSES indicates assessments of both external and internal doses recorded in Mayak PA archives and/or obtained at FIB-1 until 1999, while

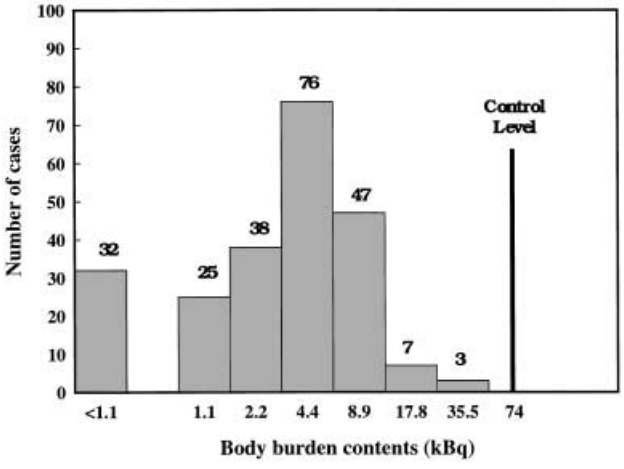

Fig. 1 Frequency distribution of body burden due to beta-activity of uranium fission (gross beta-activity for Mayak PA personnel)

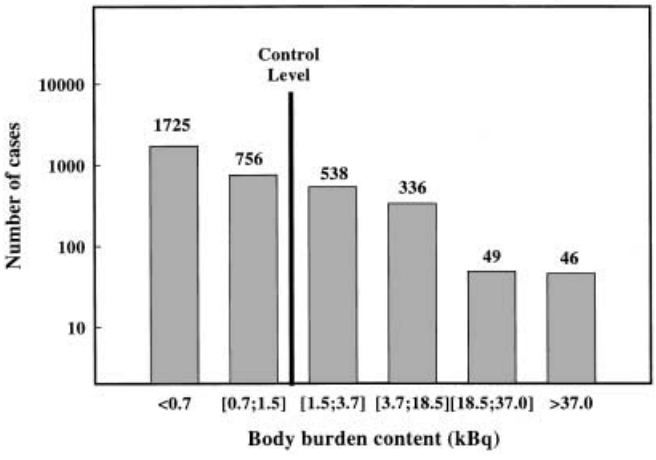

Fig. 2 Frequency distribution for the plutonium body burden of the Mayak cohort. Assessment results are given for 31 December 1998 for those alive then, otherwise for the date of death

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[1] In 2001, the name of the institute has been changed to Southern Urals Biophysics Institute.

Table 1 Dosimeter performances used at Mayak PA

Dosimeter type	Application period	Compensating filter	Error[a]	Energy range
IFK (film)	1948–1953	Without filter	±30%	0.4–3.0 MeV
IFK+Pb (film)	1954–1960	Lead 0.75 mm	Not yet determined	Not yet determined
IFKU (film)	1961–1992	Lead 0.75 mm and 0.5 mm aluminium	±30%; Additional error due to energy dependence of sensitivity ±20%	0.1–3.0 MeV
DDG-01 (thermoluminescent)	1993 up to now	Lead and aluminium 1 g/cm^2	±15%; Additional error due to energy dependence of sensitivity ±20%	0.05–3.0 MeV

[a] expressed by standard deviations.

the term NEW_DOSES indicates dose assessments obtained by applying first corrections. The Mayak PA and FIB-1 archives contain information on external doses for about 100,000 persons including Mayak PA workers, personnel of contracting agencies, military personnel and those persons who were involved in the elimination of the consequences of the radiation accident at Mayak PA in 1957 and on internal doses for 8,694 Mayak PA workers. At FIB-1 the data are completely computerized, and at Mayak PA about 50% of the information is entered into the database.

External exposure

Four types of personal dosimeters have been used to monitor external exposures of Mayak workers (Table 1). In the early period (1948–1953) the IFK film dosimeter was used, its main limitation, however, was that it was not suitable for appropriately measuring photons with energies below 400 keV. The dosimeters subsequently applied were equipped with compensating filters continuously reducing the lower limit of the energy range.

The highest gamma-radiation doses of the Mayak PA workers were recorded during the start-up and adaptation phase of the reactor and the radiochemical plants (1948–1952). Average annual values of tissue equivalent doses in free air due to external exposures amounted to 1 Gy (Fig. 3), and maximum individual doses were up to 8 Gy per year. At that time individual control was conducted at every shift and sometimes every operation. By 1953–1955, maximum doses dropped to 1 Gy per year, and average doses to 0.1 Gy per year. Since 1968 at the radiochemical plant and since 1974 at the reactor plant, individual annual doses have not exceeded 50 mGy and the average annual level of external exposure has been about several mGy. These historical dose records are reconsidered in the frame of several international programs, which include the major tasks listed below.

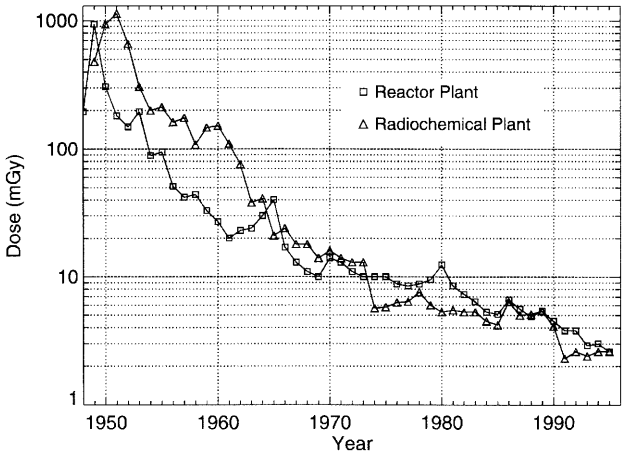

Fig. 3 Average annual external doses of reactor and radiochemical workers (Fig. reproduced from [14])

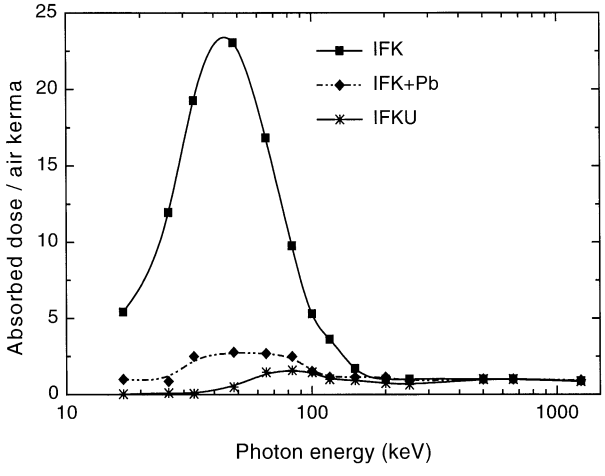

Fig. 4 Energy dependence of dose sensitivity for personal film dosimeters IFK, IFK+Pb and IFKU

Dosimeter characteristics

This work includes the reconstruction of both old dosimeter types (with assessment of their sensitivity to low energy photons, electrons and anisotropic exposures) and calibration conditions (including comparative measurements with all dosimeter types under real plant conditions).

The energy dependence of the dosimeter sensitivity as presented in Fig. 4 has been determined by measurements in the Laboratory of Secondary Standards at GSF-National Research Center of Environment and Health in Neuherberg, Germany. Accordingly, the greatest uncertainty in in-

Table 2 Correction factors K defined by the ratio of historically recorded doses to improved dose estimates (ranges given in the third column apply to different work places in the production divisions; $\overline{K}$ is the average value and $\delta\overline{K}$ its standard deviation)

Production division	Dosimeter type	Range of correction factors (K)	$\overline{K}$	$\delta\overline{K}$ (%)
Radiochemical production: Entrance and intermediate divisions	IFK	1.10–1.46	1.28	15
	IFK+Pb	0.94–1.01	0.98	4
	IFKU	0.89–0.94	0.92	3
Radiochemical production: Ending divisions	IFK	1.02–2.8	1.91	47
	IFK+Pb	0.96–1.21	1.09	11
	IFKU	0.88–1.02	0.95	7.4
Plutonium production: Radiochemical divisions	IFK	1.02–2.8	1.91	47
	IFK+Pb	0.96–1.21	1.09	11
	IFKU	0.88–1.02	0.95	7.4
Plutonium production: Chemical and metallurgical divisions	IFK	1.13–1.73	1.42	21
	IFK+Pb	0.84–0.96	0.90	4
	IFKU	0.79–0.85	0.82	3.5
Reactor production: Central hall	IFK	1.01–1.65	1.33	22
	IFK+Pb	1.0–1.05	1.025	2.5
	IFKU	0.95–1.01	0.98	3.1
Reactor production: Other divisions, besides central hall	IFK	1.10–1.46	1.28	15
	IFK+Pb	0.94–1.01	0.98	4
	IFKU	0.89–0.94	0.92	3

dividual dose assessment has to be expected for the period from 1948 to 1953, when dosimeters were used that did not have filters to compensate for the energy dependence on sensitivity. The IFK dosimeter has a very large sensitivity to photons with energies below 150 keV, with a maximum value of 23 at 50 keV, if calibrated with a ^{137}Cs source.

Reconstruction of radiation fields at the work place

It was necessary to reconstruct the source term parameters in the 1950s–1980s, to perform radiation transport calculations for different production areas of the work place with the aim of determining photon fluences (including their dependence on energy and direction) and to make measurements of photon spectra with high purity germanium detectors [2] in order to validate the simulation calculations for current working conditions.

An analysis of historical records showed that there is practically no information on the energy distribution of the photon fields at different Mayak PA working areas during the initial period of operation. Recently performed photon transport calculations and measurements yielded spectra at a few current Mayak PA production areas [2]. Calculations and measurements agree well and show that above the storage site for spent fuel rods, photons with energies below 400 keV contribute 25–30% to the absorbed dose in air whereas photons with energies below 150 keV contribute 5–10%. The corresponding spectral contributions at the shore of lake Karachai, used for waste disposal, were 85% and 40%, respectively.

Improvements to estimates for doses due to external photon exposures

The results on the sensitivity of detectors and on the radiation fields at work places are combined to improve the historical dose estimates and to assess uncertainties. Ratios of historical dose records and improved dose estimates are denoted by a correction factor K (Table 2). The spread of the factor K in Table 2 depends on the distance from the source, on the presence or absence of a biological shield, and on its thickness and composition. In total, calculations were performed for 120 reconstructed photon spectra.

During the early years of Mayak PA operation (1948–1953, when IFK dosimeters were used), the gamma doses for the personnel were mostly overestimated, in some work areas by a factor of 2.8. After the introduction of IFKU dosimeters in 1960, the individual doses were sometimes slightly underestimated due to dosimeter insensitivity to photons with energies below 100 keV (Fig. 4).

Reconstruction of external doses due to exposures not monitored by individual dosimeters

This study includes dose assessment for exposures to both neutron and beta radiation. Since 1983, the doses due to neutron fields at Mayak PA have been routinely controlled with track dosimeters. A first approach to the reconstruction of neutron doses was performed and the results for certain production areas and working groups were expressed in the form of ratios $K_n=D_n/D_\gamma$, where D_n and D_γ are tissue equivalent doses in free air due to neutron or gamma exposures, respectively. A radiation weighting factor for neutrons of 12 was assumed, based on measured spectra and on values for the specific equivalent dose per neutron fluence according to the Russian Radiation Safety Standards [3]. The values given in these standards are similar to those published in NCRP Publication No. 38 [4].

First estimates for K_n varied from 0.02 to 0.45. The maximum contribution of neutrons to the individual dose (for a

limited number of Mayak PA personnel) was observed at terminal production areas of the radiochemical and plutonium plants. At the reactor plant a substantial part of the workers were exposed to neutrons. However, there was only a minor contribution of neutrons to their overall dose.

Under routine conditions, beta exposures did not contribute significantly to the radiation doses of the workers in the working areas of Mayak PA. However, for accident situations, the reconstruction of electron fields is necessary in order to be able to derive their contribution to the blackening of the film dosimeters.

Comparison of dose estimates obtained by different ethods

The dose estimates based on the occupational dosimetry records are now being compared with estimates based on EPR measurements of absorbed dose in tooth enamel and on fluorescence in-situ hybridisation (FISH) measurements of chromosome aberrations.

EPR measurements were made on 62 teeth which had been extracted from Mayak PA workers for medical reasons [5,6]. Because of the different dosimeter characteristics, the workers of Mayak PA were subdivided into two groups, depending on whether they received more than 50% of their dose before or after 1 January 1961.

For the first group (24 workers), the EPR results on absorbed dose in enamel were highly correlated with estimates of dose in air that were based on film badge readings, on average they were 25% lower (Fig. 5). These results confirm that during the first period of operation the exposure of Mayak PA workers was generally overestimated by the historical records. For the second group (38 workers), the correlation (r=0.61) was not as high as in the first group. The average ratio of the two dose estimates was 1.1, which corresponds to the ratio of the absorbed doses in enamel and in air. It is a future task to take angular and energy characteristics of the radiation field into account to allow a more precise validation of the existing dose estimates with the help of EPR measurements.

Comparing the dose estimates obtained by FISH measurements of chromosome aberrations with the historical dose records for 69 Mayak PA workers showed that the yield of aberrations among the workers was lower than expected according to historical dose records and a calibration curve. The difference between these two dose estimates varied considerably among the individual workers [7]. According to this study and the much better correlation of EPR measurements with the historical dose records, FISH measurements of chromosome aberrations are of limited use in the validation of dose estimates for Mayak PA workers.

Internal exposure

Since 1970, the plutonium content has been measured in samples of diurnal urine at the biophysical laboratory of FIB-1. To derive internal doses from the results of these measurements (which need to be continued), the following tasks had to be accomplished.

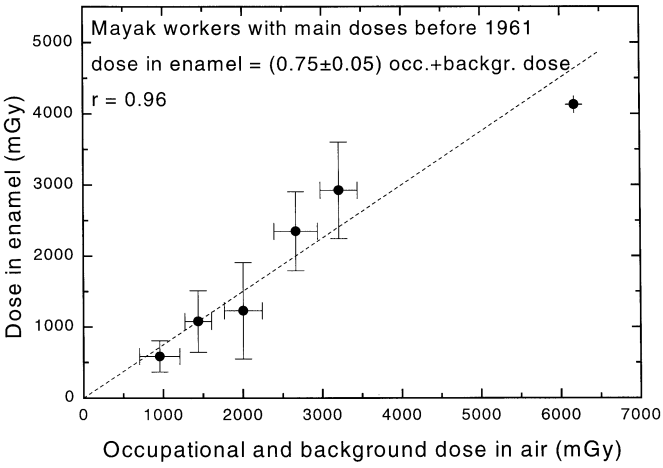

Fig. 5 Absorbed dose in tooth-enamel of workers at Mayak in comparison with an assessment of the natural and man-made background radiation dose in Ozyorsk plus occupational doses that predominantly occurred before 1961 (points and error bars represent mean values and standard deviations of results for five workers each, the value for a dose exceeding 4,000 mGy representing one worker)

Radiochemical analyses of organ/tissue specimens from autopsied Mayak PA workers (“Autopsy” program)

Radiochemical analyses of organ/tissue specimens from autopsied Mayak PA workers have been carried out with the aim of obtaining information on plutonium distribution in the human body. By 2001, the “Autopsy” database included data on 1206 post-mortem measurements of former Mayak PA workers. Inter-laboratory comparisons with the U.S. Transuranium and Uranium Registries (USTUR) showed an equivalence of the procedures for sampling, radiochemical analysis and radionuclide measurement in biological material. This made it feasible to combine the data obtained in each of the two institutes for joint data processing [8, 9].

Development of a classification system for plutonium industrial aerosols

Plutonium inhaled with industrial aerosols is classified according to physical and chemical properties. The transportability index was chosen as a main criterion that predetermines the plutonium behavior in the respiratory tract [10]. Transportability is determined by measuring the plutonium fraction that penetrates a semi-permeable membrane during 2 days of dialysis in Ringer’s solution. Table 3 shows that the plutonium content in lung and pulmonary lymph nodes of autopsied workers correlates highly with the transportability index. The issue of the correlation between the transportability index and the dispersion ability of plutonium industrial aerosols remains unsolved.

Table 3 Transportability of industrial aerosols at Mayak PA work places and the percentage of post-mortem plutonium body burden remaining in the respiratory tract (geometric mean values and geometric standard deviations are given in parentheses)

Workplace	Reprocessing of uranium fuel	Plutonium fuel production (workplace 1)	Plutonium fuel production (workplace 2)
Transportability (%)	3.0 (1.7)	1.0 (3.0)	0.3 (1.6)
Number of cases	223	261	59
Lung content[a] (%)	2.6 (2.2)	7.5 (2.4)	25 (1.8)
Lung and lymph nodes content[a] (%)	4.2 (2.4)	15 (2.4)	50 (1.5)

[a] Percent of total body burden.

Development of a model for plutonium excretion

Based on the data obtained at FIB-1 for 25 persons, Durbin's model [11] for the late period after plutonium inhalation was modified [12]. This model employs a superposition of five exponential terms to predict the plutonium content e_u in the urine of a healthy person at a time t (in days) after a plutonium content q appears in the blood:

$$e_{\mathrm{u}} = q \times 10^{-4}(41\exp(-0.56t) + 12\exp(-0.126t) + 1.3\exp(-0.0165t) + 0.3\exp(-0.00231t) + 0.13\exp(-0.00002t)).$$

The corresponding expression for the plutonium content e_f in feces is:

$$e_{\mathrm{f}} = q \times 10^{-4}(60\exp(-0.35t) + 16\exp(-0.105t) + 1.2\exp(-0.0124t) + 0.2\exp(-0.00182t) + 0.052\exp(-0.00002t)).$$

There were no gender differences in plutonium metabolism.

Information on the radiation environment at various work areas and autopsy data were used to approximate the plutonium deposition rate in the human body by a function exponentially decreasing with time.

Development of a lung clearance model

The structure of the FIB-1 lung clearance model was developed based on recommendations of ICRP-30 [13]. The major characteristic of the model compared to those previously used is the presence of a component representing a plutonium fraction that is fixed within the lung tissue [14].

Plutonium measurements in urine ("Bioassay" program)

By 2001, the "Bioassay" database included information on about 80,000 bioassays for 8,694 Mayak PA workers with corresponding estimates of the plutonium body burden.

Although the FIB-1 model predicts the plutonium body burden satisfactorily, its drawbacks are evident, the main one being that the clearance of the respiratory tract towards the throat (particle clearance) is not taken into account. Neglecting this clearance leads to an underestimation of the lung dose particularly in the early phase after intake, if the dose assessment is based on a measurement of the plutonium content in urine and a related content in the lungs at a later time after the intake. Therefore, the FIB-1 model was revised by adapting the ICRP-66 lung clearance model to the actual data on the metabolism of plutonium industrial compounds in the Mayak PA workers [15].

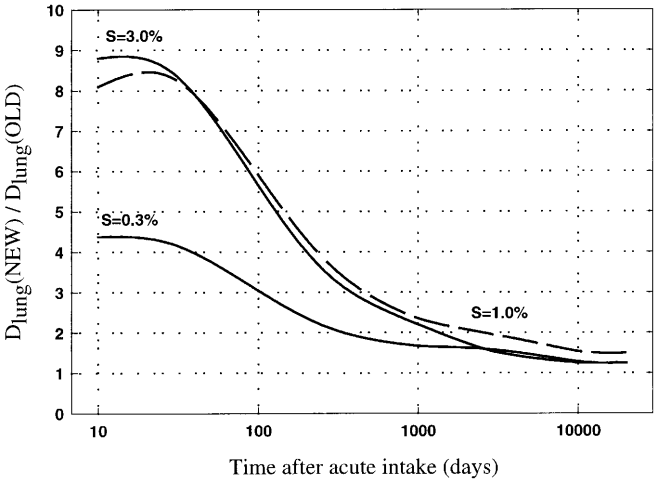

Fig. 6 Ratio of accumulated absorbed doses in the lung after acute inhalation of plutonium in forms with different transportability

Based on available data for nuclide urine excretion as well as on exposure histories for persons from the studied cohort, plutonium burdens and internal doses were calculated in accordance with the new model. A preliminary comparison (Fig. 6) indicates that on average "new" doses are higher than "old" ones by a factor of 1.3 and immediately after intake these dose assessments differ by an order of magnitude [16].

Concerning the sub-cohort of Mayak PA workers for whom measurements of plutonium content in urine have been performed, the highest effective radiation doses were recorded for Mayak PA workers hired between 1948 and 1953. Assuming a radiation weighting factor for Pu of 20 [17], an average effective dose of 5.56 Sv was obtained for this period. More than 60% of the dose was due to internally deposited plutonium (Table 4). The annual radiation doses decreased until the early 1980s by 2 orders of magnitude as a result of measures taken to improve working conditions.

Future improvements to the internal dosimetry system will involve an adaptation of the systemic model to fit actual data obtained by FIB-1 scientists and a refinement

Table 4 Dose estimates for the sub-cohort of Mayak PA workers for whom measurements of the plutonium content in urine have been made (*1* "DOSES_OLD", *2* "DOSES_NEW")

Year of beginning of contact	Methods	1948–1953	1954–1958	1959–1963	1964–1972	1973–1982
Average absorbed lung dose [Gy]						
Internal	1	0.49	0.12	0.07	0.023	–
	2	0.54	0.15	0.09	0.036	0.013
Average effective dose [Sv]						
Total	1	5.16	1.39	0.64	0.207	–
	2	5.56	1.67	0.80	0.270	0.116
By Internal	1	3.25	0.76	0.41	0.140	–
	2	3.68	1.04	0.57	0.200	0.074
By External	1	1.91	0.63	0.23	0.067	–
	2	1.88	0.63	0.23	0.070	0.042

of individual doses to organs of main deposition by both the in-depth analysis of occupational exposure histories and the inclusion of metabolic modifying factors (such as state of health, smoking status, or age).

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